



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

In re Application of

BINGEL et al.

Serial No. 09/701,658

Filed: November 30, 2000

For: METALLOCENE MONOHALIDES

Art Unit: 1713

Examiner: R. Lee

Honorable Commissioner of
Patents and Trademarks
Washington, D.C. 20231

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BRIEF ON APPEAL

Sir:

This Appeal is from the Examiner's Final Rejection of July 31, 2002.

REAL PARTY IN INTEREST

The real party in interest is Targor GmbH of Fed. Rep. Germany. Reel 011445,
Frame 0695, recorded on November 30, 2000.

RELATED APPEALS AND INTERFERENCES

To the best of the undersigned's knowledge, there are no related appeals or
interferences within the meaning of 37 CFR 1.192(c).

STATUS OF CLAIMS

Claims 9 to 14 stand finally rejected under 35 U.S.C. 103(a).

STATUS OF AMENDMENTS

An amendment after final was mailed on January 3, 2003. This amendment,

which was not entered, was in response to an objection of claim 9 by the examiner.

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An amendment accompanying this Brief has been filed to correct some inadvertent errors in claim 12. It is assumed in the Brief that this amendment will be entered since it eliminates potential issues.

SUMMARY OF INVENTION

The invention is directed to particular bridged metallocene monohalides having at least one indenyl ring attached to the bridging group where the metallocenes have certain oxygen or sulfur to carbon moieties attached to the transition metal. These oxygen and sulfur to carbon moieties are defined in claims 9 and 12 by "Y" and "R³".

These metallocenes have the advantage over their dihalide counterparts of being readily soluble and at the same time show catalyst activities in polymerizations at least as good as the corresponding metallocene dihalides (page 1 of specification, lines 37-41).

ISSUES

Is the subject matter of claims 9-14 unpatentable over Tsutsui et al. (U.S. 5,795,838) in view of Repo et al. (J. Organomet. Chem. 541(1997)363-366)?

It is assumed that the examiner did not withdraw the Repo reference since the Advisory Action of February 5, 2003 did not specifically do so. Note item 3 on PTO - 300 form and item 5 in the Advisory Action. The reference to 35USC102(b) in the Advisory Action is assumed to be a typographical error.

GROUPING OF CLAIMS

Claims 11-12 do not stand or fall with claims 9-14 and will be argued separately.

ARGUMENT

The examiner's position appears to be that Tsutsui does not disclose compounds wherein a halogen and an aryloxy group are attached to the transition metal. The examiner points out that formula (Ia) in claim 14 of Tsutsui states that X^1 and X^2 may be the same or differing and maybe an oxygen containing group or a sulfur containing group. In the same claims formula(Ic) states that X^3 and X^4 (equivalent positions and X^1 and X^2 in formula (Ia)) may be C_6-C_{10} aryloxy. The examiner then concludes that it would be obvious to one of ordinary skill in the art to derive such a compound because Tsutsui states that X^1 and X^2 may be different from one another and each formula discloses use of both type of ligands. They would be expected to be useful catalyst precursors according to the examiner because they come with the disclosure of the reference.

In the Advisory Action the examiner further points out that compounds in col. 10 at lines 48-49 and 56 have the formula $Cp_2M(X)(OR)$ where Cp is cyclopentadienyl and X is chloride and R is methyl or ethyl are known, further showing that such monohalogen complexes are not outside the teaching of the reference.

In the final rejection based on Tsutsui in view of Repo, the examiner states that Tsutsui discloses the use of bridged bis(indenyl) based metallocenes containing one C_6-C_{10} aryloxy group; however the reference does not show any example of the aryloxy group. Since Repo discloses monohalide zirconocene compounds containing two

specific aryloxy groups, one of ordinary skill in the art having both references would find it obvious to use the aryloxy groups or Repo et al. to make the compound of the claims.

The examiner should be reversed because any possible *prima facie* case of obviousness has been rebutted by the evidence of record.

It is surprising and unexpected that the claimed metallocenes show at least comparable polymerization activity to their dihalide analogs while being so readily soluble at the same time (page 1, lines 37-41). More specifically, the metallocenes of the invention have a significantly better solubility in inert organic solvents; as explained on page 7 of the instant application, 1st full paragraph; the concentration of the claimed metallocenes in organic solvents is at least doubled, and even 8 times higher the one of the corresponding bichloride compounds (see the solubility comparative trial in Examples 1-4 and 6). This property is very important at the industrial level in the purification of racemic metallocenes by crystallization techniques. Moreover, the metallocenes of the instant invention display a better crystallization behavior from inert organic solvents, which notably improve their purification process (page 7, lines 7-16 of the application).

This much greater solubility is a factor that the examiner has not accounted for in his determination of obviousness. The examiner affirms that the fact that monohalide complexes are significantly more soluble than the dichloride derivatives is not terribly surprising since there are several well-established ways to improve solubility of metallocene in an inert organic solvent and one involves modifying the compound itself. We believe that this reason cannot be the basis for a rebuttal to applicants arguments

since it is not corroborated by evidence. While the examiner may rely on logic and sound scientific principles in support of a rejection under § 103 the examiner must provide some evidentiary basis for the existence and meaning of the scientific principle relied on. See *In re* Grose, 592 F. 2d 1161, 1167-68, 201 USPQ 57, 63 (CCPA 1979). Further, merely modifying a compound obviously does not inherently make it more soluble.

In the Advisory Action the examiner stated:

"The examiner maintains the notion that solubility of the compounds shown by the Applicants is not unexpected nor surprising based on the fact that the aryloxy substituent possesses two branched aliphatic groups. Not everything qualifies as a surprising result. If one were to believe the Applicants, the mere fact that compounds of the invention can be made at all would be "surprising" (see specification, page 17, lines 1-3)."

It is not strictly the much greater solubility that is unexpected but rather the combination of having much greater solubility than their dihalide analogs and at least comparable activity that is unexpected.

The record also establishes the greater or at least the comparable polymerization activity of the claimed metallocenes over their dihalide counterparts.

The closest prior art to claim 9 would appear to be Tsutsui since it is the only reference that includes a structurally similar metallocene species having three of the same groups present in the same relative position, i.e., indenyl group, bridge and chlorine. Repo, by contrast, has only two of the same groups, i.e., an alkylaryloxy and

chlorine radical. The claimed compounds, however, show unexpected results over species of Tsutsui having the three common ligands. Thus one of the preferred species of Tsutsui, dimethylsilanediybis(2-methyl-4,5-benzoindeny)-zirconium dichloride shown in Example 13 and also shown in the Comparative Example on page 33 of the specification, unexpectedly produced inferior catalytic activity when used as a catalyst precursor in the polymerization of propylene as opposed to one of the claimed species, dimethylsilane diylbis(2-methyl-4,5-benzoindeny)-zirconium monochloride mono-2,4-di-tert-butylphenoxide). The metallocenes only differ as to the presence of the alkyloryloxy group while the polymerizations were back-to-back runs. The catalyst activities were 5300 g of pp/g of catalyst x h for the claimed metallocene and 4600g of pp/g of catalyst x h for the dihalide preferred species of Tsutsui. While the examiner described these results as "comparable" (page 3 of Advisory Action), the 15% improvement in catalytic activity is especially significant since the metallocene dihalides are generally considered to be industrially favored and are the obvious preferred species of the art of record, e.g., Tsutsui (note species in columns 9, 10 and 27 - 33 and all of the working examples 1-17). Further the claimed monohalide was more than 36 times as soluble in toluene (page 33, lines 1-6) than its dihalide counterpart, which is a great advantage as explained above. Thus, this showing is considered to be significant and unexpected.

The other art of record in the rejection, Repo also confirms the unexpectedness of the results shown in the specification. In three out of four comparative polymerization runs in Repo, the aryloxy containing metallocenes were less active than

their dihalide counterpart. Thus, Repo and even Tsutsui teach away from using the claimed monohalide metallocenes. This evidence of teaching away from the claimed invention must be considered in determining unobviousness under 35 USC103(a). See MPEP, Sect. 2141.03, Prior Art Must Be Considered In its Entirety, Including Disclosures That Teach Away From The Claims. Also see *In Re May* 197 USPQ 601.

The examiner, however, even believes that unexpected results have been shown, i.e., "What is unexpected and surprising, then, is the fact that present compounds display comparable activity compared to their dihalide analogues" (page 3 of Advisory Action). This is what the application stated (page 1, lines 37-41 and page 29, lines 18-22). Thus it is believed that any possible *prima facie* case of obviousness has been rebutted by the evidence of record and as confirmed by the examiner.

Claims 11 and 12 are even further removed from the reference since they are directed to metallocenes having two indenyl groups. Claim 12 is further restricted to two substituted indenyl groups.

CONCLUSION

The examiner should be reversed because the record as a whole rebuts any possible *prima facie* case of obviousness established by the applied prior art.

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BINGEL et al.

Serial No. 09/701,658

Respectfully submitted,

KEIL & WEINKAUF

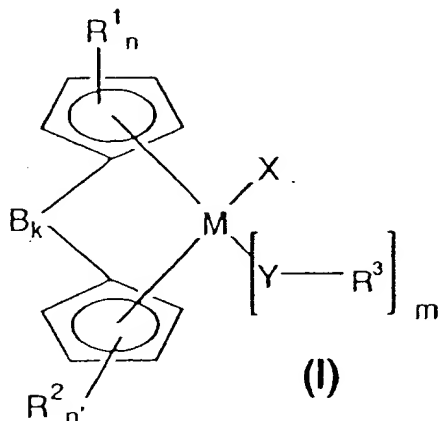
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APPENDIX

9. A compound of the formula (I),



where

M is a metal of transition group III, IV, V or VI of the Periodic Table of the Elements,

R^1 are identical or different and are each a radical $Si(R^{12})_3$, where R^{12} are identical or different and are each a hydrogen atom or a C_1 - C_{40} -group or R^1 is a C_1 - C_{30} -group, or two or more radicals R^1 may be connected to one another in such a way that the radicals R^1 and the atoms of the cyclopentadienyl ring which connect them form a C_4 - C_{24} -ring system which may in turn be substituted,

R^2 are identical or different and are each a radical $Si(R^{12})_3$, where R^{12} are identical or different and are each a hydrogen atom or a C_1 - C_{40} -group, or R^2 is a C_1 - C_{30} -group, or two or more radicals R^2 may be connected to one another in such a way that the radicals R^2 and the atoms of the cyclopentadienyl ring which connect them form a C_4 - C_{24} -ring system which may in turn be substituted,

R^3 are identical or different and are each a C_2 - C_{25} -alkenyl, C_3 - C_{15} -alkylalkenyl, C_5 - C_{24} -heteroaryl, C_7 - C_{30} -arylalkyl, C_7 - C_{30} -alkylaryl, fluorinated C_1 - C_{25} -alkyl, fluorinated C_6 - C_{24} -aryl, fluorinated C_7 - C_{30} -arylalkyl or fluorinated C_7 - C_{30} -alkylaryl,

X is a halogen atom,

Y is oxygen or sulfur,

n is from 0 to 4,

n' is from 0 to 4,

m is from 1 to 3,

k is 1,

B is a bridging structural element between the two cyclopentadienyl rings and

one or both cyclopentadienyl rings are substituted in such a way that they form an indenyl ring.

10. A compound as claimed in claim 9, wherein

M is Ti, Zr or Hf,

R^1 are identical or different and are each a radical $Si(R^{12})_3$, where R^{12} are identical or different and are each a hydrogen atom a C_1 - C_{20} -alkyl, C_1 - C_{10} -fluoroalkyl, C_1 - C_{10} -alkoxy, C_6 - C_{10} -aryl, C_6 - C_{10} -fluoroaryl, C_6 - C_{10} -aryloxy, C_2 - C_{10} -alkenyl, or R^1 is C_1 - C_{25} -alkyl, C_2 - C_{25} -alkenyl, C_3 - C_{15} -alkylalkenyl, C_6 - C_{24} -aryl,

C₅-C₂₄-heteroaryl, C₇-C₃₀-arylalkyl, C₇-C₃₀-alkylaryl, fluorinated C₁-C₂₅-alkyl, fluorinated C₆-C₂₄-aryl, fluorinated C₇-C₃₀-arylalkyl, fluorinated C₇-C₃₀-alkylaryl, or C₁-C₁₂-alkoxy, or two or more radicals R¹ may be connected to one another in such a way that the radicals R¹ and the atoms of the cyclopentadienyl ring which connect them form a C₄-C₂₄-ring system which may in turn be substituted,

R² are identical or different and are each a radical Si(R¹²)₃, where R¹² are identical or different and are each a hydrogen atom a C₁-C₂₀-alkyl, C₁-C₁₀-fluoroalkyl, C₁-C₁₀-alkoxy, C₆-C₁₀-aryl, C₆-C₁₀-fluoroaryl, C₆-C₁₀-aryoxy, C₂-C₁₀-alkenyl, or R² is C₁-C₂₅-alkyl, C₂-C₂₅-alkenyl, C₃-C₁₅-alkylalkenyl, C₆-C₂₄-aryl, C₅-C₂₄-heteroaryl, C₇-C₃₀-arylalkyl, C₇-C₃₀-alkylaryl, fluorinated C₁-C₂₅-alkyl, fluorinated C₆-C₂₄-aryl, fluorinated C₇-C₃₀-arylalkyl, fluorinated C₇-C₃₀-alkylaryl, or C₁-C₁₂-alkoxy, or two or more radicals R² may be connected to one another in such a way that the radicals R² and the atoms of the cyclopentadienyl ring which connect them form a C₄-C₂₄-ring system which may in turn be substituted,

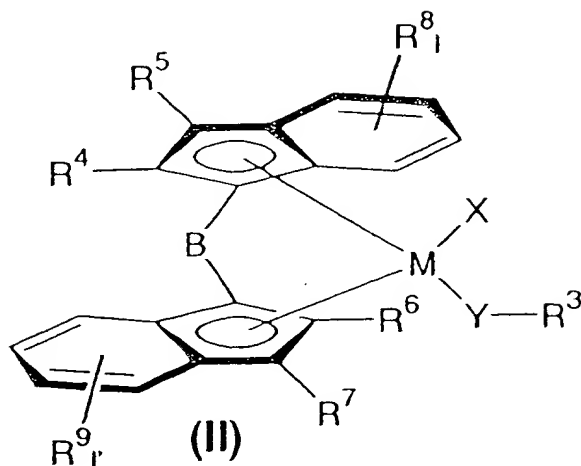
X is chlorine

Y is oxygen or sulfur,

m is 1 and

one or both cyclopentadienyl rings are substituted in such a way that they form an indenyl ring which is substituted.

11. A compound of the formula (II)



where

M is Ti, Zr or Hf,

R³ is isopropyl, tert-butyl, cyclohexyl or octyl, a C₅-C₂₄-heteroaryl, C₇-C₃₀-arylalkyl, C₇-C₃₀-alkylaryl, fluorinated C₆-C₂₄-aryl, fluorinated C₇-C₃₀-arylalkyl, or fluorinated C₇-C₃₀-alkylaryl

R⁴, R⁶ are identical or different and are each a hydrogen atom or a C₁-C₂₀-group,

R⁵, R⁷ are identical or different and are each a hydrogen atom or a C₁-C₂₀-group,

R⁸, R⁹ are identical or different and are each a hydrogen atom, a halogen atom or a C₁-C₂₀-group, and two radicals R⁸ or R⁹ may form a monocyclic or polycyclic ring system which may in turn be substituted,

X is a halogen atom,

Y is oxygen or sulfur,

I, I' are identical or different and are each an integer from zero to 4,

B is a bridging structural element between the two indenyl radicals.

12. A compound as claimed in claim 11, wherein, in the formula (II),
- M is zirconium,
- R⁴, R⁶ are identical or different and are each a hydrogen atom, a C₁-C₁₈-alkyl, C₂-C₁₀-alkenyl, C₃-C₁₅-alkylalkenyl, C₆-C₁₈-aryl, C₅-C₁₈-heteroaryl, C₇-C₂₀-arylalkyl, C₇-C₂₀-alkylaryl, fluorinated C₁-C₁₂-alkyl, fluorinated C₆-C₁₈-aryl, fluorinated C₇-C₂₀-arylalkyl or fluorinated C₇-C₂₀-alkylaryl,
- R⁸, R⁹ are identical or different and are each a hydrogen atom, a halogen atom a linear or branched C₁-C₁₈-alkyl group, C₂-C₁₀-alkenyl, C₃-C₁₅-alkylalkenyl, a C₆-C₁₈-aryl group which may be substituted, C₅-C₁₈-heteroaryl, C₇-C₂₀-arylalkyl, C₇-C₂₀-alkylaryl, fluorinated C₁-C₁₂-alkyl, fluorinated C₆-C₁₈-aryl, fluorinated C₇-C₂₀-arylalkyl or fluorinated C₇-C₂₀-alkylaryl, and two radicals R⁸ or R⁹ may form a monocyclic or polycyclic ring system which in turn may be substituted,
- X is chlorine,
- Y is oxygen or sulfur,
- I, I' are identical or different and are each 1 or 2.
13. A catalyst comprising at least one compound as claimed in claim 9 and a support and, optionally, a cocatalyst.
14. A process for preparing a polyolefin which comprises polymerizing an olefinic monomer in the presence of a catalyst as claimed in claim 13.